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Environmental Radiation Safety: Plutonium/Soil Interactions For Plutonium Particles in Soil

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Prepared for
Office of Advanced Nuclear Systems and Projects
Space and Terrestrial Systems Division
for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

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ABSTRACT

The goal of this project is to provide information useful in estimating hazards related to resuspension characteristics and subsequent aerodynamic behavior of aerosols from a mixture of soil and $^{238}PuO_2$. Experiments were carried out to determine whether simple models, used to predict the total activity concentration of resuspended particles, need to be modified to account for changes in the $^{238}PuO_2$ activity distribution on resuspended particles due to aging of the soil mixture under humid or dry conditions.

A literature search revealed that one model, based on the suspension factors, S_f, may be a useful predictor of hazard reduction irrespective of site. The relation is: $S_f = 10^{-4}e^{-0.15\sqrt{t}} + 10^{-9}$, where t is time in days and S_f = the ratio of the air concentration of resuspended material (Ci/m^3) divided by the surface deposition (Ci/m^2). Our experiments demonstrated little or no change in the activity of resuspended particles following humid or dry aging of the soil-$^{238}PuO_2$ mixture. Additional terms for activity distribution changes should not be needed for the simple resuspension hazard model.

INTRODUCTION

BACKGROUND

The goal of this project is to provide information useful in estimating hazards related to the use of a pure refractory oxide of ^{238}Pu as a power source in some of the space vehicles to be launched during the next few years. Although the sources are designed and built to withstand re-entry into the earth's atmosphere, and to impact with the earth's surface without releasing any plutonium, the possibility that such an event might produce aerosols composed of soil and $^{238}PuO_2$ cannot be absolutely excluded. Should $^{238}PuO_2$ become mixed in a finely divided form with soil, the resuspension characteristics and subsequent aerodynamic behavior of the aerosols would be major factors in determining the fate of the plutonium in the environment. Humidity cycling and time in the ground would be expected to change the amount and aerodynamic distribution of the resuspended fraction of soil and $^{238}PuO_2$ particles.

At the Nevada Test site and at Rocky Flats, Colorado, there are sites where plutonium has been released as an aerosol, mixed with the soil, and aged. Measurements have been made on the content and size distribution of plutonium particles in soil samples and in resuspended aerosol samples collected on filters.[1-10] At Rocky Flats, the reported respirable fraction of plutonium dioxide in samples of the soil mixture ranges from less than 10%[3] to as high as 20 to 40%[2] of the total activity present (dpm/g). The experimental technique used to measure the respirable fraction of plutonium may have caused the difference in the results.[3,10,11] In fact, the increase in the respirable soil fraction is significant (5 to 10 times as much), depending on whether the soil sample remains dry or has water added during the separation technique, since particle surface forces are less in water. Hayden[3]

and Tamura[10] both estimate that about 80% of the plutonium can be dissociated from the larger soil particles into respirable-sized particles if enough dispersive energy is supplied to overcome the surface forces.

The resuspended particles consist of soil, or plutonium, or soil-plutonium particles that have been released from large particles due to collision. This collision also occurs in saltation during a sandstorm, or during transfer of an airborne parent particle into a raindrop.[12,13,14] Wind intensity and soil texture influence the number of soil particles resuspended. With the same sand bombardment conditions, fine-textured soils produce more airborne particles in the <10-μm range than do coarse-textured soils. This may be a partial explanation for differences in field samples. Production of resuspended soil particles of <10-μm diameter increased with increased horizontal sand flux at Rocky Flats,[15] but the movement of soil in saltation was not significant at the site in Nevada.[5] Plutonium particles resuspended above Rocky Flats in July of 1973 were attached to host soil particles. The distribution of activity (dpm/g) was uniform except for rare hot particles.[7] Resuspension rates, RR, for undisturbed soils at this site increased with windspeed, W, ($RR \sim W^{6.5}$), and decreased as soil moisture increased.[8]

Exact models predicting the resuspension of particles observed in the field are difficult to develop.[13,14,16] Furthermore, those models based on resuspension rates[14] may provide more detail than is necessary for a risk analysis such as that in the Overall Safety Manual.[17] The relation between soil content and potential lung burden from resuspended particles is difficult to predict accurately.[1-4,9,10] Resuspension rates measured at the Nevada and Colorado sites were low, on the order of 10^{-10} to 10^{-8} fractions resuspended/sec.[8] At the Nevada site, 20 years after plutonium release, resuspension continued to occur from undisturbed soil. Airborne concentrations were 1/120 of the maximum permissible airborne concentration (MPC) for yearly exposure to

respirable plutonium, 0.6 pCi/ m^3,[18] for samples at 500 feet from ground zero, the point of initial release, and were approximately equal to the MPC at ground zero. At Rocky Flats, for a soil concentration of 3 nCi/g (6890 dpm/g), the maximum observed respirable plutonium concentration from resuspended particles was 3.7×10^{-15} Ci/m^3, which is about 1/169 of the MPC. The data suggest that the inhalation risk from resuspension of plutonium from soil mixtures may be modeled by a simple function decreasing to near-zero risk in as little as 5 years. Anspaugh, et al.[13] report such a model based on the resuspension factor (S_f); the ratio of the air concentration of resuspended material (Ci/m^3) divided by the surface deposition (Ci/m^2). In their example, the airborne concentration decreased by a factor of 100 to 1000 during the first 100 hours following the explosion. Thereafter, the airborne concentration, represented by the resuspension factor, decreased according to the following equation (corrected from their text, which contained a printing error):

$$S_f(m^{-1}) = 10^{-4}e^{-0.15\sqrt{t}} + 10^{-9}, \qquad (1)$$

when t = time in days from release of the source term.

Inhalation risk is determined by the fraction of the resuspended particles that is respirable. Under usual conditions of resuspension, the observed respirable mass ratio is less than the maximum that can be generated or separated from the soil-plutonium mixture. The respirable fraction of the total mass resuspended was 0.02 to 0.25 in the Rocky Flats area,[1,4,6] even for cases where the plutonium attachment (μCi/g) in the total mass resuspended was the same as the plutonium attachment (μCi/g) for the soil mixtures.[4] Tamura[10] calculated a plutonium index for redispersion to quantify the inhalation risk from resuspended plutonium particles at four different sites. The plutonium index (PI) was the product of three factors: soil activity (SA), lung deposition (LD), and resuspendable activity (RA), (PI = SA x LD x RA). The activity-to-mass ratio for each of three sieve fractions (<2, 2-5, and 5-125 μm) of the

soil and plutonium mixture is multiplied by the estimated lung deposition fraction (0.4, 0.12 and 0.03, respectively). The sum of these three ratios is the product. SA x LF. This product is multiplied by the fraction of the total activity in the less-than-25-μm sieve-size fraction, RA Significantly, he found that the plutonium index for each site was approximately 1, and that none of the four sites differed from any of the other sites by more than a factor of 2.4.

EXPERIMENTAL MODEL

High levels of respirable plutonium can be produced above a site where soil is mixed with plutonium.[10,14] The total amount of material resuspended decreases with soil moisture content, but increases with wind speed and saltation. The respirable fraction of the resuspended material would be expected to change in relation to the extent of moist or dry aging of the soil mixtures. The degree of change, which is unknown, determines the amount of additional detail needed in modeling the health risk as a function of time above a site of soil mixed with plutonium.

For the experimental model, we mixed soil and $^{238}PuO_2$, aged the mixture, and measured changes in the resuspended aerosol. The mass ratio of soil to $^{238}PuO_2$ was 10^{+5} to 1, which is 20,000 to 70,000 times the activity-to-mass ratio measured at the Rocky Flats site.[4] The plutonium attachment (μCi/g) in both the resuspended aerosol and the soil/plutonium mixture was kept equal by removing thin layers from the end surface of a packed cylinder of the soil mixture. This material was then dispersed as an aerosol. The mechanical energy transfer that takes place in saltation was modeled by directing the fluidized soil/plutonium mixture into a turbulent air stream before dispersing the dust into the test system.

Because of results reported in the literature,[19,20] the soil/plutonium mixture was aged under two conditions, dry or moist. The aging treatments used before testing are listed in Table 1 for the four soil mixtures studied: clay soil plus $^{238}PuO_2$, sandy loam soil plus $^{238}PuO_2$, clay soil only and sandy loam soil only. Once a particular batch of soil and plutonium was packed into a cylinder, treatments in the loose state could no longer be made. The 28-day humid cycle (Table 1) consisted of continuous treatment for 21 days, followed by 7 days storage in a dry atmosphere.

Four to six test runs were made following each treatment, during which total deposition, airborne concentration and particle size distribution were measured. The only significant change seen in these tests was in the clay soil-$^{238}PuO_2$ aerosol produced after continued humid treatment. The activity median aerodynamic diameter (AMAD) of the aerosol changed very little as a result of treatment (increase, 0.7%); the geometric standard deviation (GSD) increased 13%. The effect of the humid aging was to cause the $^{238}PuO_2$ particles to be slightly more uniformly distributed on the surface of the resuspended clay particles than they were before treatment. These results suggest that predictions of the total activity concentration of resuspended particles, rather than the changes in activity distribution of resuspended particles, may be the most useful in estimating reduced inhalation hazard above a site of mixed soil and plutonium.

TABLE 1. Aging Treatments Before Test: Clay Soil, Sandy Loam Soil and $^{238}PuO_2$

Soil Mixture	Initial Treatment (Aged Loose in Dish)					WDF Pack Number	Continued Treatment (Aged Packed in WDF Cup)				
	Days of Dry Cycle	28 Day Humid/Dry Cycles	Total Days	Tests: Series Number	Tests: Run Number		Days of Dry Cycle	28 Day Humid/Dry Cycles	Total Days	Tests: Series Number	Tests: Run Number
Clay Soil (S3) + $^{238}PuO_2$	1	-	1	56	1-4	1	28 + 13	7	238	56	37-42
	110	-	110	56	5-8	4	120	-	230	56	25-30
	0 + 1	4	113	56	9-12	5	28 + 1	3	233	56	31-36
	222	-	222	56	13-18	2C	-	-	-	-	-
	0 + 1	8	225	56	19-24	2D	-	-	-	-	-
Sandy Loam Soil (S1) + $^{238}PuO_2$	1	-	1	60	1-4	3	21 + 22	7	240	60	41-46
	111	-	111	60	5-10	7	121	-	232	60	29-34
	0 + 2	4	114	60	11-16	8	28 + 11	3	237	60	35-40
	223	-	223	60	17-22	6C	-	-	-	-	-
	0 + 2	8	226	60	23-28	6D	-	-	-	-	-
Clay Soil (S3)	1	-	1	61	1-5	-	-	-	-	-	-
Sandy Loam Soil	1	-	1	62	1-5	-	-	-	-	-	-

MATERIALS AND METHODS

Powders of pure plutonium oxide, sandy loam soil and clay soil were used in this experiment. The $^{238}PuO_2$ powder was sieved to a maximum particle size of 10 μm at Los Alamos Scientific Laboratories before shipment to the Pacific Northwest Laboratory in 1974. The sandy loam soil was a sample of the volcanic tuft underneath Los Alamos, NM. The clay soil was obtained from stream beds in the valley below Los Alamos. The mass median diameters and geometric standard deviations of the sandy loam soil and clay soil were measured by sedimentation and sieving.[20] The mass median diameters were 57 μm (sandy loam) and 1.1 μm (clay), with geometric standard deviations of 4.4 and 6.0, respectively. Soils were stored dry or under 93% relative humidity. Humidity in the closed container was maintained by use of an ammonium sulfate solution.

THE WIND TUNNEL

The low-speed wind tunnel used for this study was built for a study of plutonium uptake in plants from foliar deposition.[21,22] The tunnel, constructed of stainless steel, is 178 cm long; the inlet end has a square cross-section, 20.3 cm on each side; the center section is 30.5 cm on each side. The transition between the two sections of the tunnel is shown in Figure 1. Plants and equipment can be positioned through a file-cabinet-type drawer located in the center section of the tunnel.

To provide double containment, the entire wind tunnel is enclosed in a 2.4 x 0.8 x 0.8-m Plexiglas glove box. In operation, a negative pressure differential exists between the glove box and the room, as well as between the tunnel and the glove box.

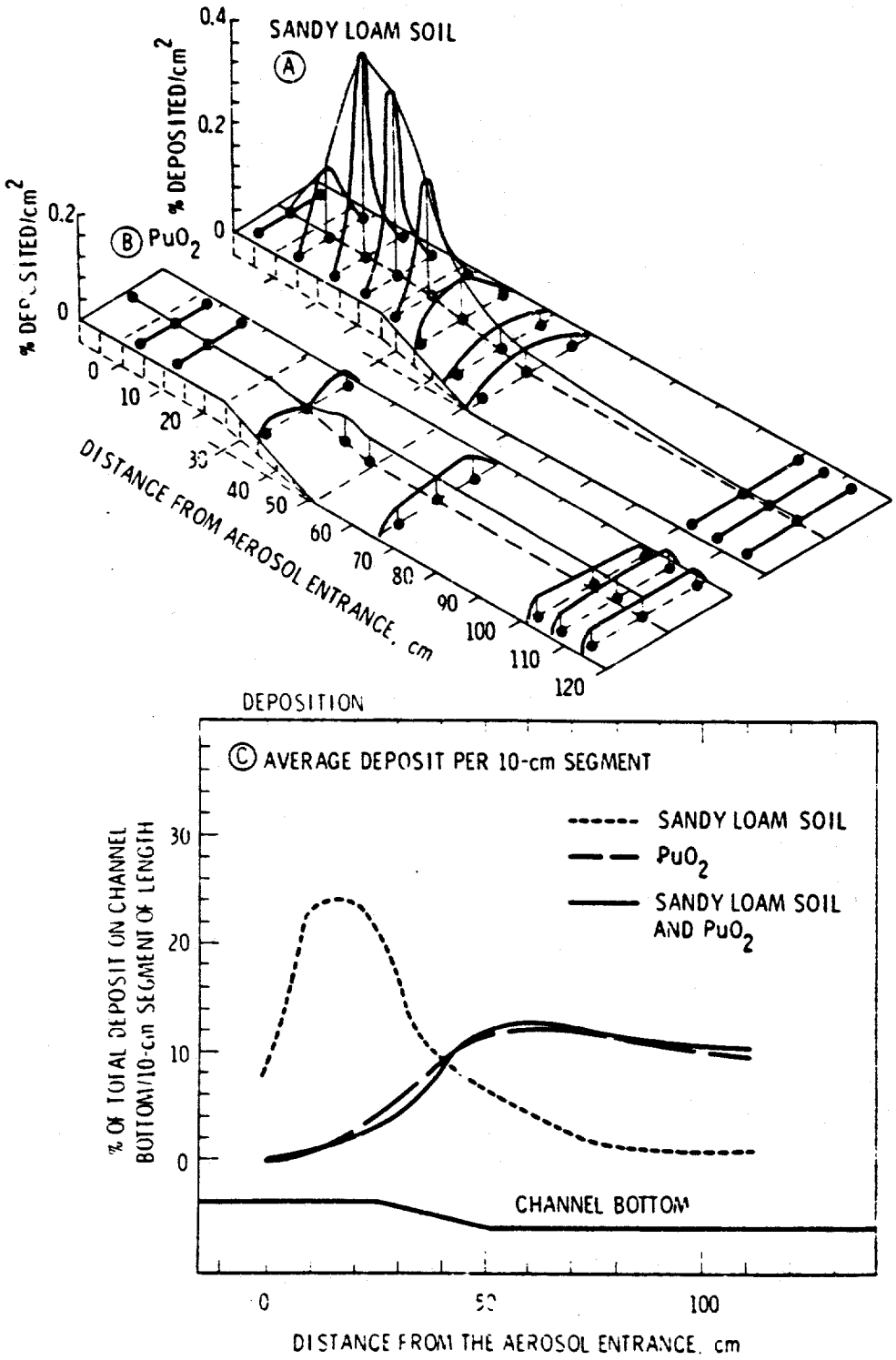

FIGURE 1. Filter Distribution Pattern and Deposited Activity.

SAMPLING: TOTAL MASS (OR TOTAL ACTIVITY) AIRBORNE

Isokinetic filter-paper samples were taken at midchannel, at locations 57, 81, 123 and 139 cm from the outlet of the aerosol mixing nozzle. Sample flow rates of 0.15 ℓ/min were drawn through 25-mm filters.

SAMPLING: PARTICLE SIZE, AMAD AND MMAD

The AMAD of the mixed soil/$^{238}PuO_2$ aerosol was measured with a Mercer cascade impactor (MCI), operated at 0.6 ℓ/min. The stages of the impactor were gamma-counted in the same manner as the filter samples. The MMAD of the soil aerosol was measured with an Andersen cascade impactor (ACI), operated at 18 ℓ/min. Each stage of the impactor was weighed. All impactor samples were taken approximately at midchannel, 88 cm from the end of the aerosol mixing nozzle. All MCI samples were taken isokinetically. The ACI samples were taken as close to isokinetic conditions as possible. Only 23% of the total flow through this impactor was from the sampling nozzle; the rest was supplied by dilution air.[20]

SAMPLING: PARTICLE SIZE, AR SIZING

The size distribution of the $^{238}PuO_2$ particles was measured using autoradiography.[25,26] Samples of the sedimented dust were collected on a 25-mm filter, positioned on the floor of the wind tunnel 103 cm from the end of the aerosol mixing nozzle. Samples of the airborne dust were collected on a 25-mm filter positioned at midchannel, at locations 57 cm and 139 cm from the end of the aerosol mixing nozzle. The filters, each containing no more than 2 nCi of activity, were sent to Dr. M. W. Nathans (LFE, Environmental Analysis Laboratories, 2030 Wright Avenue, Richmond, CA 94804) for autoradiographic analysis.

Soil particles associated with $^{238}PuO_2$ particles were sized on selected filters by matching the fields on the autoradiographic film and on the light microscope slide.[26]

RESULTS

The experimental approach was to generate an aerosol of $^{238}PuO_2$ mixed with clay or sandy loam soil into the wind tunnel and measure the amount deposited, the amount airborne, the aerodynamic size distribution and the autoradiographic size distribution of the plutonium particles. Except for the change in air velocity in the wind tunnel, all other aerosol generation, mixing and sampling conditions were kept as constant as possible so that any changes observed in the aerosol could be attributed to pre-treatment of the soil-plutonium mixture by dry and/or humid atmospheres (Table 1).

AEROSOL GENERATOR OUTPUT

Total mass output of the aerosol generator was measured by weighing the WDF dust feed cylinder before and after each run ("total amount generated," Table 2). The $^{238}PuO_2$ output was calculated by measuring the specific activity of the soil/plutonium mixture ("estimated from generator output," Table 2). The series and run number for each row of Table 2 (Column 1) correspond to those given in Table 1. The experiment identification number (same nomenclature as presented in our previous report)[27] corresponds to the information summarized in Table 1:

S3 = clay soil

S1 = sandy loam soil

L = loose; soil or soil/plutonium mixture stored in the loose state

P = packed; soil or soil/plutonium mixture stored in the packed state in the WDF dust feed cylinder cup

D = dry storage conditions

W = wet or humid storage conditions

TABLE 2. Summary of the Soil-Plutonium Aging Experiments

	EXPERIMENT NUMBER AND IDENTIFICATION	AIR VELOCITY IN THE CHANNEL (cm/sec)	GENERATION TIME Δt (min)	TOTAL AMOUNT GENERATED: AIRBORNE + DEPOSIT (mCu[a])	ESTIMATED FROM GENERATOR OUTPUT (g)	ESTIMATED FROM GENERATOR OUTPUT (mCi)	MASS OF SOIL IN THE GENERATOR OUTLET (mg)	AIRBORNE CONCENTRATION AT 110 cm (nCi/ℓ[b])	DISTRIBUTION % AIRBORNE	% DEPOSITED	% OTHER	AEROSOL DISTRIBUTION SAMPLED AT 97 cm: CRITICAL χ^2 9.2 AMAD	GSD	χ^2	% 10μm	% 6μm
56 1	S1D1 1d1	0.9	20	0.047	1.2	6.03	154	4.5	9.5	82.1	8.4	17.5	3.1	0.5		17.7
56 2	"	0.9	20	0.050	1.2	0.03	168	5.5	10.9	83.1	6.0	18.7	3.3	0.4		16.5
56 3	"	22.4	20	0.136	1.2	0.03	128	4.0	73.5	24.5	1.9	14.9	3.0	11.0		25.0
56 4	"	22.4	20	0.048	1.2	0.03	130	1.5	89.8	10.6	3.6	50.2	6.1	5.6		13.8
56 5	S3LD110.11d4	0.9	20	0.085	1.3	0.10	177	11.0	13.0	64.9	22.1	12.3	2.7	1.0		22.4
56 6	"	0.9	20	0.049	1.3	0.10	173	1.7	1.5	84.6	11.9	10.5	2.8	1.4		27.0
56 7	"	22.4	20	0.028	1.3	0.10	133	0.8	68.0	12.6	19.4	4.9	3.8	31.9		65.6
56 8	"	22.4	20	0.096	1.3	0.10	143	0.9	23.4	30.0	46.5	29.1	4.2	2.8		16.5
56 9	S3LIW21D7ηD1 113d5	0.9	20	0.044	1.4	0.08	176	2.1	5.3	75.2	19.5	10.6	2.4	0.2		27.0
56 9'		0.9	11									12.4[d]	2.6	0.4		24.0
56 10	"	0.9	20	0.043	1.4	0.08	189	3.6	7.4	65.9	26.7	12.4	3.1	0.3		16.9
56 11	"	22.4	20	0.036	1.4	0.08	146	0.2	14.8	62.9	22.9	34.2	4.1	1.6		12.0
56 12	"	22.4	20	0.094	1.4	0.08	140	1.2	92.1	55.8	12.1	256.4	6.8	0.44		2.4
56 13	S3LD222 222d2(	0.9	20	0.087	1.3	0.06	185	9.0	10.4	59.5	30.1	12.3	2.8	1.4		25.0
56 14	"	0.9	20									19.1[d]	3.4	0.6		16.5
56 15	"	0.9	20	0.050	1.3	0.06	184	6.0	11.9	73.9	14.2	21.2	3.4	0.3		16.0
56 16	"	22.4	20	0.112	1.3	0.06	161	2.6	57.8	22.5	19.7	68.7	4.9	0.9		7.0
56 17	"	22.4	20									30.1[d]	3.5	3.0		12.5
56 18	"	22.4	20	0.020	1.3	0.06	130	0.2[e]	23.0	52.1	24.9	7.8	2.0	21.1		59.0
56 19	S3LIW21D7ηD1 225d2D	0.9	20	0.063	1.3	0.07	159	1.2	5.1	65.9	29.0	15.5	2.9	0.2		14.5
56 20	"	0.9	20									16.4[d]	3.5	1.2		22.0
56 21	"	0.9	20	0.039	1.3	0.07	192	1.9	4.6	81.6	13.8	10.6	2.8	1.3		33.0
56 22	"	21.0	20	0.025	1.3	0.07	167	0.2	22.6	57.5	19.7	49.6	6.5	2.6		14.0
56 23	"	22.4	20									21.8[d]	3.1	5.0		17.0
56 24	"	21.0	20	0.039	1.3	0.07	135	0.05	3.3	40.2	56.5	3.0	2.6	55.1		69.0
56 25	S3r8PD120 230d4	0.9	20	0.040	1.3	0.10	183	5.5	11.4	50.5	35.7	26.2	3.5	3		16.5
56 26	"	0.9	20									11.9[d]	2.7	0.7		27.0
56 27	"	0.9	20	0.039	1.3	0.10	190	2.7	6.9	73.3	19.8	18.8	3.9	0.9		19.0
56 28	"	22.4	20	0.063	1.3	0.10	166	0.4	14.6	63.0	22.4	42.9	5.9	5.1		17.5
56 29	"	22.4	20									53.9[d]	4.9	0.9		5.0
56 30	"	22.4	20	0.044	1.3	0.10	154	0.9	52.9	26.3	20.9	8.7	2.8	58.6		74.0
56 31	S3r12+PD28IW21D7ηD1 235d5	0.9	20	0.027	1.4	0.08	161	1.5	5.5	75.1	19.3	19.0	3.9	3.0		33.0
56 32	"	0.9	20									9.2[d]	2.2	1.1		15.0
56 33	"	0.9	20	0.041	1.4	0.08	202	1.3	4.3	54.1	41.6	15.6	3.2	0.1		11.0
56 34	"	22.4	20	0.088	1.4	0.08	166	0.5[e]	12.9	44.4	42.7	9.9	5.1	26.5		35.0
56 35	"	22.4	20									65.1[d]	9.0	9.4		10.0
56 36	"	22.4	20	0.024	1.4	0.08	146	0.6	57.3	15.6	27.1	4.9	4.2	45.0		51.0
56 37	S3r4+PD28IW21D7ηD1 238d1	0.9	20	0.015	1.3	0.02	140	1.0	6.8	57.7	35.5	1.6	3.6	2.0		74.0
56 38	"	0.9	20									7.3[d]	2.5	1.7		33.0
56 39	"	0.9	20	0.028	1.3	0.02	181	1.3	4.7	67.4	27.9	16.7	4.1	0.2		17.0
56 40	"	22.4	20	0.040	1.3	0.02	159	0.96	59.6	32.2	8.2	16.7	4.2	7.7		36.0
56 41	"	22.4	20									114.7[d]	48.2	9.3		9.0
56 42	"	22.4	20	0.030	1.3	0.02	153	0.23[e]	19.2	60.3	20.5	6.1	4.0	62.9		86.0
60 1	S1D1 1d3	0.9	20	0.152	1.7	0.28	192	20.2	13.2	66.8	20.0	6.0	2.5	2.1		30.0
60 2	"	0.9	20	0.170	1.7	0.28	142	21.6	12.7	66.4	20.9	5.9	2.3	2.0		30.0
60 3	"	22.4	20	0.149	1.7	0.28	152	2.1	35.3	46.2	18.5	19.8	3.8	0.9		17.0
60 4	"	22.4	20	0.158	1.7	0.28	143	2.7	32.7	43.6	13.7	10.1	2.6	0.3		30.0
60 5	1LD111.111d7	0.9	20	0.204	1.5	0.26	219	27.2	13.3	66.1	20.6	5.7	2.1	1.6		54.0
60 6	"	0.9	21		1.5	0.27						5.1[d]	2.2	0.4		56.0
60 7	"	0.9	20	0.195	1.5	0.26	206	24.4	17.5	68.2	19.5	5.7	2.2	0.1		52.0
60 8	"	22.4	20	0.164	1.5	0.26	132	2.5[e]	37.6	44.8	17.6	8.3	2.6	0.8		38.0
60 9	"	22.4	20		1.5	0.26						6.9[d]	1.9	9.4		49.0
60 10	"	22.4	20	0.124	1.5	0.26	142	1.8	35.1	48.1	16.8	11.9	2.8	0.8		27.0

[a] UNIT IS mCi UNLESS OTHERWISE INDICATED

[b] UNIT IS nCi/ℓ UNLESS OTHERWISE INDICATED

[c] CRITICAL VALUE OF χ^2 = 10.2

[d] SAMPLED AT 130 cm

[e] AVERAGE OF LAST SAMPLES

TABLE 2 (continued).

EXPERIMENT NUMBER AND IDENTIFICATION	AIR VELOCITY IN THE CHANNEL (cm/sec)	GENERATION TIME, ΔT (min)	TOTAL AMOUNT GENERATED: AIRBORNE + DEPOSIT (mCi)[a]	TOTAL AMOUNT GENERATED: ESTIMATED FROM GENERATOR OUTPUT (g)	TOTAL AMOUNT GENERATED: ESTIMATED FROM GENERATOR OUTPUT (mCi)	MASS OF SOIL IN THE GENERATOR OUTLET (mg)	AIRBORNE CONCENTRATION AT 110 cm (nCi/g)[b]	DISTRIBUTION % AIRBORNE	DISTRIBUTION % DEPOSITED	DISTRIBUTION % OTHER	AEROSOL DISTRIBUTION SAMPLED AT 82 cm: CRITICAL x^2 9.2 AMAD	GSD	Σx^2	% < 10 μm	% < 6 μm
60-11 S1E1W21D7$_4$D2 114d8	0.9	20	0.169	1.5	0.27	216	26.8	13.9	66.5	19.6	7.0	2.5	3.7		40.0
60-12 "	0.9	20	"	1.5	0.27		"	"	"	"	5.1[d]	2.3	3.2		54.0
60-13 "	0.9	20	0.197	1.5	0.27	176	32.0	16.2	69.2	14.6	6.4	2.2	0.3		47.0
60-14 "	22.4	20	0.104	1.5	1.27	139	2.0	48.1	29.7	22.2	13.1	3.2	1.3		24.0
60-15 "	22.4	20	"	1.5	0.27		"	"	"	"	17.8[d]	2.6	0.7		14.0
60-16 "	22.4	20	0.108	1.5	0.27	139	4.5	51.9	29.1	19.0	8.2	2.7	1.8		39.0
60-17 S1E1D223.223ddC	0.9	20	0.201	1.5	0.28	199	18.0	9.0	72.9	18.1	6.4	2.5	2.0		48.0
60-18 "	0.9	20	"	1.5	0.28		"	"	"	"	4.7[d]	2.2	0.6		67.0
60-19 "	0.9	20	0.204	1.5	0.28	165	16.2	7.9	74.7	17.4	5.8	2.3	1.2		57.0
60-20 "	22.4	20	0.095	1.5	0.28	102	1.7	45.0	33.2	21.8	32.7	4.2	0.2		19.0
60-21 "	22.4	20	"	1.5	0.28		"	"	"	"	9.4[d]	2.7	3.8		39.0
60-22 "	22.4	20	0.184	1.5	0.28	136	3.4	46.1	35.3	18.6	13.4	2.9	0.7		23.0
60-23 S1E1W21D7$_8$D2 226ddD	0.9	20	0.170	1.5	0.32	188	15.0	8.8	72.1	19.1	4.8	2.2	4.8		62.0
60-24 "	0.9	20	"	1.5	0.32		"	"	"	"	4.5[d]	2.1	0.7		66.0
60-25 "	0.9	20	0.158	1.5	0.32	190	16.0	10.1	68.5	21.4	5.4	2.2	0.1		62.0
60-26 "	22.4	20	0.168	1.5	0.32	155	3.0	44.6	39.3	16.1	10.1[d]	6.9	8.1		32.0
60-27 "	22.4	20	"	1.5	0.32		"	"	"	"	23.9[d]	2.7	1.2		32.0
60-28 "	22.4	20	0.172	1.5	0.32	153	2.9	42.2	43.8	14.0	8.1	2.7	1.3		42.0
60-29 S1r10+ PD121.232d7	0.9	20	0.120	1.4	0.25	163	15.0	12.5	68.5	19.0	5.9	2.3	0.5		48.0
60-30 "	0.9	20	"	1.4	0.25		"	"	"	"	5.9[d]	2.4	0.4		55.0
60-31 "	0.9	20	0.110	1.4	0.25	169	16.0	15.4	60.0	24.6	6.5	2.5	1.1		49.0
60-32 "	22.4	20	0.134	1.4	0.25	131	2.1	39.2	44.1	16.7	7.3	2.8	3.5		44.0
60-33 "	22.4	20	"	1.4	0.25		"	"	"	"	7.6[d]	2.4	1.0		44.0
60-34 "	22.4	20	0.149	1.4	0.25	125	2.2	37.0	49.5	13.5	18.1	3.3	0.3		18.5
60-35 S1r16+ PD28W21D7$_3$D11.237d8	0.9	20	0.140	1.6	0.28	151	11.0	7.8	74.7	17.5	5.6	2.2	0.6		53.0
60-36 "	0.9	20	"	1.6	0.28		"	"	"	"	4.5[d]	2.1	1.4		66.0
60-37 "	0.9	20	0.122	1.6	0.28	149	13.5	11.0	65.8	23.2	5.3	2.1	1.7		61.0
60-38 "	22.4	20	0.152	1.6	0.28	113	3.3	54.2	32.8	13.0	22.9	4.1	1.5		17.5
60-39 "	22.4	20	"	1.6	0.28		"	"	"	"	13.0[d]	3.1	3.5		26.0
60-40 "	22.4	20	0.094	1.6	0.28	121	1.5	40.1	40.8	19.1	10.7	2.9	2.0		38.0
60-41 S1r4+ PD21HW21D7$_7$D22 240d3	0.9	20	0.234	1.6	0.27	201	19.5	8.1	75.3	16.6	6.7	2.2	0.1		46.0
60-42 "	0.9	20	"	1.6	0.27		"	"	"	"	4.9[d]	2.1	1.3		67.0
60-43 "	0.9	20	0.114	1.6	0.27	156	14.0	12.2	67.4	20.4	5.5	2.2	1.0		55.0
60-44 "	22.4	20	0.111	1.6	0.27	131	1.5	33.8	47.5	18.7	16.0	3.7	1.8		27.0
60-45 "	22.4	20	"	1.6	0.27		"	"	"	"	8.1[e]	2.4	1.3		83.0
60-46 "	22.4	20	0.173	1.6	0.27	130	1.5	21.7	47.6	30.7	19.4	3.3	0.9		17.0
61-1 S3+ HOH	0.9	97	5.9 g	5.4		890	230.0 μg/g	18.9	66.1	15.0	3.5	2.6	20.8[c]	85.0	
61-2 "	22.4	296	21.0 g	19.0		1805	21.5 μg/g	37.9	53.5	8.6	4.5	2.8	32.1[c]	78.0	
61-3 "	0.9	185	11.2 g	11.9		1530	254.0 μg/g	26.5	65.5	13.6	3.4	2.6	20.5[c]	88.0	
61-4 "	0.9	95	5.9 g	6.0		870	243.0 μg/g	19.5	65.8	14.7	3.2	2.3	19.5[c]	90.0	
61-5 "	0.9	95	4.7 g	6.0		867	222.0 μg/g	22.3	59.3	18.4	3.3	2.3	20.3[c]	93.0	
62-1 S1+ HOH	0.9	170	12.2 g	12.5		1869	62.0 μg/g	4.3	80.3	15.4	5.3	2.2	35.9[c]	77.5	
62-2 "	22.4	300	21.0 g	21.1		2490	7.5 μg/g	13.4	74.8	11.8	11.4	2.7	7.1[c]	45.0	
62-3 "	0.9	175	14.0 g	13.0		1955	69.0 μg/g	4.3	81.7	14.0	4.7	2.4	20.3[c]	80.0	
62-4 "	0.9	175	11.4 g	12.8		1936	63.0 μg/g	4.8	78.2	17.0	4.8	2.3	14.0[c]	82.0	
62-5 "	0.9	175	11.7 g	12.4		1868	49.0 μg/g	3.7	80.3	16.0	5.4	2.3	16.3[c]	78.0	

[a] UNIT IS mCi UNLESS OTHERWISE INDICATED

[b] UNIT IS nCi/g UNLESS OTHERWISE INDICATED

[c] CRITICAL VALUE OF x^2 10.2

[d] SAMPLED AT 130 cm

[e] AVERAGE OF LAST 3 SAMPLES

d = days

rN = run #N.

The numbers following L,P,D,W, and those preceding d, indicate days of treatment. The subscript indicates the number of times an aging treatment was performed. The numbers and letters following d indicate the WDF cup number used to generate the aerosol. For example: Run 56-37 (Table 2) has the identification number, S3r4 + PD28(W21D7)$_7$D13, 238d1. Reading from left to right, this can be interpreted as: "Clay soil from run #56-4 was generated again after being aged under packed dry conditions for 28 days followed by 7 humid cycles. Each cycle consisted of 21 days of humid conditions followed by 7 days of dry conditions. The 7 humid cycles were then followed by 13 days of dry atmosphere. This soil/plutonium mixture was aged for a total of 238 days. WDF cup #1 was used for the packed storage and for aerosol generation."

SAMPLING: TOTAL MASS (OR TOTAL ACTIVITY) DEPOSITED

Total mass (or activity) deposited in the wind tunnel during each experimental run can be calculated from three columns in Table 2: "Total amount generated; airborne + deposit," "Distribution; % Deposited," and "Distribution; % other." The numbers in the "% Deposited" column were obtained from the filter-paper samples on the wind-tunnel floor. The "% Other" column in Table 2 was obtained from the total mass (or activity) deposited in the aerosol mixing nozzle during each run. This mass (or activity), plus that estimated to be on the tunnel bottom, were added to the total airborne mass passing through the tunnel to obtain the "Total Airborne + Deposit," and the corresponding percentages.

The mass (or activity) deposited on each filter (at positions shown in Figure 1) is converted to deposition rate (mg or nCi/cm^2/min). The total

amount deposited was calculated by averaging the deposition rates across the width of the channel. The average deposition rate was plotted as a function of the distance from the end of the aerosol mixing nozzle. This curve was broken up into a histogram with 10-cm-long units at the base. The total amount deposited on the wind tunnel bottom was estimated by multiplying the area under the curve by the generation time. In Figure 1(c) this curve has been normalized so that the results of several experiments could be plotted on one graph.

SAMPLING: TOTAL MASS (OR TOTAL ACTIVITY) AIRBORNE

The airborne concentration at 110 cm from the aerosol inlet is also given in Table 2. The total airborne mass (or activity) passing through the tunnel was then calculated, based on the generation time and air velocity in the channel (Table 2), plus the channel cross-sectional area, 930 cm^2.

SAMPLING: PARTICLE SIZE, AMAD

The aerosol size data consisted of the total amount of material (or activity) collected on the eight stages of the cascade impactor. An effective cut-off diameter was calculated for each stage, and the resulting cumulative distribution fit to a log normal distribution.[20] The aerodynamic size distribution for each aerosol is listed in Table 2 under the heading, "Aerosol distribution sampled at 82 cm; AMAD; GSD; $\Sigma\chi^2$; % <10 pm and % <6 μm." The chi-square values, "$\Sigma\chi^2$," are a measure of how close the cascade impactor data fit a log-normal particle size distribution. The distribution was assumed not to be log-normal (0.1 level of significance) if the chi-square values were greater than 9.2 for the Mercer or 10.6 for the Andersen cascade impactor.

SAMPLING: PARTICLE SIZE, AR SIZING

The size distribution data from the autoradiographic analysis is given in Table 3. Data are separated according to type of sample: sedimentation samples at 103 cm, aerosol samples at 57 cm and aerosol samples at 139 cm from the aerosol inlet. Autoradiography, of course, measured only the size distribution of $^{238}PuO_2$ particles attached to the surface of soil particles. The cumulative distribution curves were visually fit to a bimodal, log-normal particle size distribution (Figure 3) because regression analyses programs were not available. We estimate that these visual-fit curves reproduce the data within the accuracy of the sampling and analysis technique. The characteristic values describing the biomodal distributions are given in the "$^{238}PuO_2$ Particle Sizing" columns of Table 3.

The results of light microscope sizing of soil particles near, or associated with, plutonium particles are given in the columns of Table 3 labeled "Soil Particle Sizing." The percent of $^{238}PuO_2$ particles not associated with any soil particle is given in the last column.

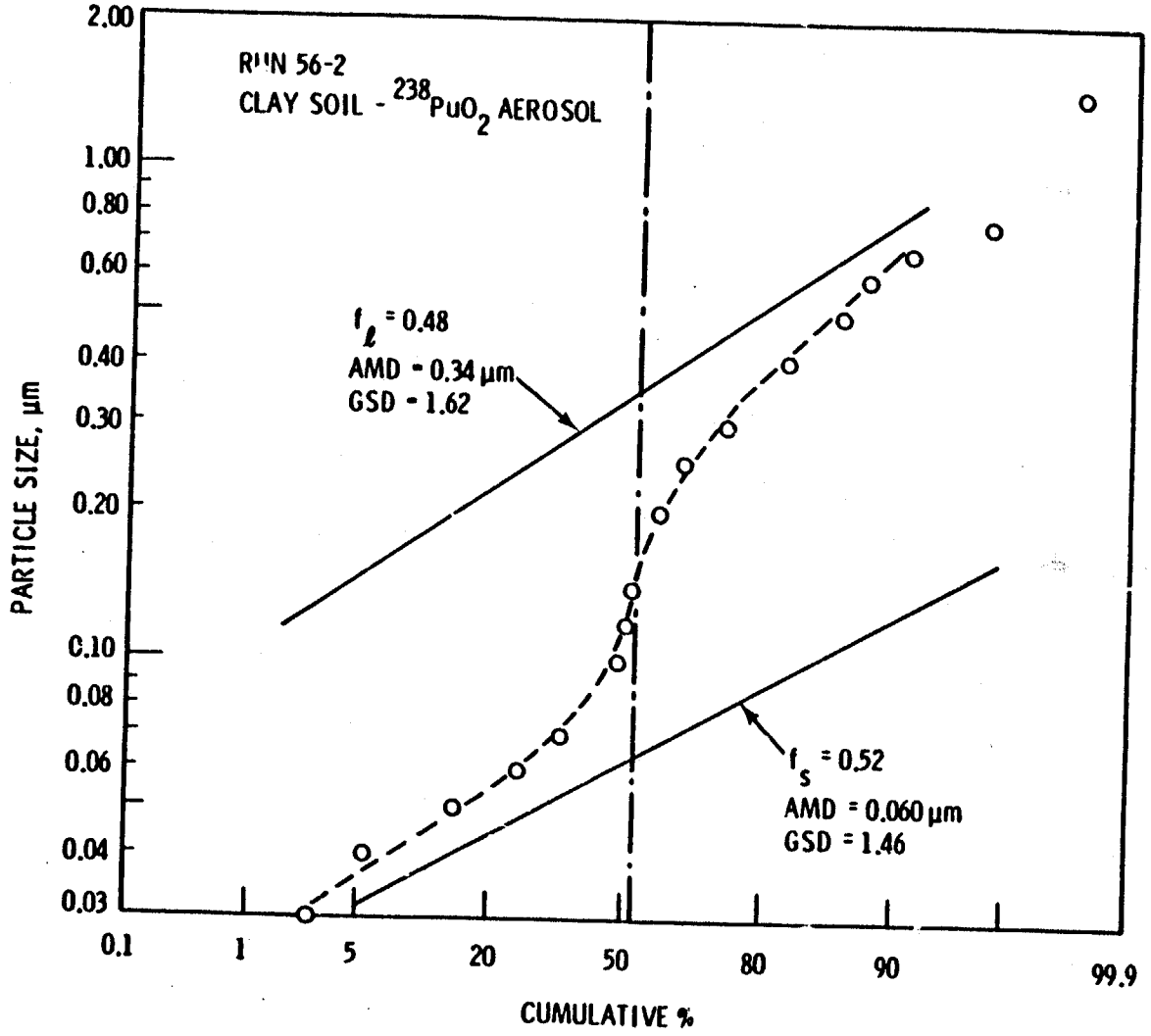

FIGURE 3. Autoradiograph Sizing; Run 56-2, Clay Soil/$^{238}PuO_2$ Aged Under Dry Conditions and Sampled in 0.9 cm/sec Airflow, 57 cm from Aerosol Inlet to Wind Tunnel (AMD = Activity Median Diameter, GSD = Geometric Standard Deviation).

TABLE 3. Autoradiographic Measurement of $^{238}PuO_2$ Particle Size Distribution

Type of Sample	Location of Sample (cm from inlet)	Air Velocity in Wind Tunnel cm/sec)	Soil	Aging Regime	Run Number
Sedimentation	103	0.9	Sandy loam	dry	60-1
				humid	60-12
			Clay	dry	56-1
				dry	56-5
				humid	56-10
Aerosol	57	0.9	Sandy loam	dry	60-6
				humid	60-11+12
			Clay	dry	56-2
				dry	56-5
				humid	56-9
Aerosol	57	22.4	Sandy loam	dry	60-4
Aerosol	139	0.9	Sandy loam	dry	60-1
				dry	60-6
				humid	60-11
				humid	60-12
			Clay	humid	56-9
				humid	56-10
Aerosol	139	22.4	Sandy loam	dry	60-3
				dry	60-4
				dry	60-9
				humid	60-15
			Clay	humid	56-12

Table 3. (continued)

Run Number	$^{238}PuO_2$ Particle Sizing: % of Total Count as Small Particles	Count Distribution of Small Particles: CMD[b] µm	GSD[c]	Count Distribution of Large Particles: CMD µm	GSD	N AR Sizing	Soil Particle Sizing[a]: N Soil Sizing	% Pu With Soil: <0.9 µm	0.9-2.2 µm	>2.2 µm	% Pu Without Soil
60-1	70	0.076	1.73	0.16	3.40	299	123	73	3	21	3
60-12	50	0.052	1.16	0.16	2.[illegible]	300	123	78	3	14	5
56-1	45	0.050	1.25	0.11	2.44	270	120	45	13	40	2
56-5	50	0.046	1.44	0.23	2.30	64	64	56	10	31	3
56-10	80	0.068	1.51	0.82	1.71	111	111	56	13	20	11
60-6	40	0.092	1.35	0.32	1.73	300	120	24	54	4	18
60-11+12	0	-	-	0.175	2.50	600	120	37	37	15	11
56-2	52	0.060	1.46	0.34	1.62	73	73	39	44	9	8
56-5	48	0.047	1.27	0.19	2.27	67	68	46	47	4	3
56-9	60	0.060	1.40	0.24	2.35	90	90	25	38	27	10
60-4	0	-	-	0.22	2.00	300	120	32	54	5	9
60-1	55	0.055	1.62	0.24	2.09	129	120	78	6	14	2
60-6	40	0.098	1.40	0.30	1.74	200	119	39	18	35	8
60-11	0	-	-	0.24	1.85	224	119	64	7	24	4
60-12	40	0.068	1.31	0.26	1.73	300	-	-	-	-	-
56-9	80	0.057	1.42	0.28	3.00	134	117	38	10	46	6
56-10	80	0.054	1.30	0.53	1.79	154	116	11	28	56	5
60-3	20	0.055	1.17	0.20	2.08	300	120	61	22	16	1
60-4	20	0.060	1.11	0.26	1.98	300	120	52	20	22	6
50-9	49	0.057	1.43	0.24	2.00	159	119	65	11	21	3
60-15	30	0.064	1.33	0.23	2.00	137	116	67	7	22	4
56-12	80	0 053	1.40	0.46	2.24	78	77	64	6	25	5

[a] from light microscopy of the soil particles associated with some of the plutonium particles

[b] CMS = Count Median Diameter

[c] GSD = Geometric Standard Deviation

DISCUSSION

Differences were seen in the aerodynamic diameter distribution of the resuspended soil particles. The clay soil aerosol was smaller than the sandy loam soil (MMAD = 3.35 μm, GSD = 2.45 vs MMAD = 5.05 μm, GSD = 2.30, Table 2).

The aerodynamic diameter distributions of plutonium on clay and sandy loam soil particles for all of the different aging treatments (Table 1) were subjected to a preliminary cluster analysis. The particle size distributions divided into distinct groups; soil and air velocity in the channel were the clearest criteria for the divisions. The cluster analysis was followed by a series of regression analyses in order to isolate the factors responsible for the differences in particle size distribution.

We analyzed the particle size distributions, defined by the observed proportion of $^{238}PuO_2$ activity on a stage of a cascade impactor, together with the effective cut-off diameter of that stage. A logit transformation was used to induce approximate linearity in log size.[28,29] A logit transformation of a proportion p is defined as

$$\text{logit}(p) = \log \frac{p}{1-p} .$$

The logit transformation is the inverse of the logistic distribution function F(s), where

$$F(s) = (1 + e^{-s})^{-1} .$$

Since the logistic distribution function and the normal (Gaussian) distribution function have very similar shapes, the logit transformation should induce approximate linearity for a normal distribution. Since the particle size distributions were approximately log normal in shape (Table 2), the log size should be normal, and the logit transformation should linearize the distribution. The logit transformation was used instead of inverse normal or probit

transformations primarily because of ease in computation. A model of the form

$$\text{logit}(p) = a + b \log(s)$$

was used, with a and b estimated by least squares. This model defines a probability distribution, so that the estimates of a and b can be used to obtain estimates of the GSD, σ, and the AMAD, μ. The relationship between these two sets of parameters is

$$\sigma = \exp\left(\frac{\pi}{b\sqrt{3}}\right)$$

$$\mu = \exp\left(-\frac{a}{b}\right).$$

The significance of an experimental factor (such as aging treatment in Table 1), or a combination of factors, was assessed using regression analysis. The significance test is carried out by fitting a single regression equation to the data pooled over similar experimental factors, followed by fitting separate regression equations to each group corresponding to a separate treatment or factor. The reduction in the residual sum of squares due to grouping provided a basis for an F-test of experimental factor significance.

Initial tests of significance were carried out for the two factors, air velocity in the channel and type of soil mixed with $^{238}PuO_2$. The first test compared all four soil x velocity groups. Four additional tests compared velocity within soil and soil within velocity. All tests resulted in highly significant ($P < 0.0001$) F statistics. The higher velocities tended to yield larger particle sizes and greater GSDs, as did the clay soil (Table 4).

Because of the extreme differences in values between the soil x velocity groups, subsequent test were performed within groups. The following sequence of tests was carried out for each soil in the low-velocity (0.9 cm/sec) groups:

TABLE 4. Aerodynamic Diameter Size Distributions for Velocity and Soil Groups (No Separation According to Aging Treatment)

Soil	Wind Tunnel Velocity (cm/sec)	AMAD μm	GSD
S1, Sandy loam	low (0.9)	4.8	1.82
$+^{238}PuO_2$	High (22.9)	9.7	2.40
S3, Clay	Low (0.9)	9.5	2.21
$+^{238}PuO_2$	High (22.4)	19.9	3.73

groups: Test 1 compared overall difference between treatment regimens. Test 2 compared data from regimens using loose, dry aging to data from those using loose, humid aging. If Test 2 showed no significant difference, all data from the loose, dry aging regimens were pooled and compared to those from the packed, dry regimen (Test 3) and to packed, humid regimens (Test 4). The results of these tests are summarized in Table 5.

None of the test results were significant for soil 1, the sandy loam soil. In Test 1, the overall treatment difference was highly significant ($P < 0.0001$) for soil 3, the clay soil. The difference in Test 2, dry versus humid aging, was not significant, nor was that of Test 3, loose versus dry, packed aging. However, in Test 4, loose versus packed, humid aging, the difference was highly significant ($P < 0.0001$) for soil 3. Distribution parameters for the treatment groups in Tests 3 and 4 are given in Table 6.

TABLE 6. Aerodynamic Diameter Size Distributions for S3, Clay Soil/$^{238}PuO_2$ Mixture (Air Velocity in Wind Tunnel = 0.9 cm/sec)

Treatment	AMAD μm	GSD
Loose aging	9.4	2.15
Packed dry aging	10.1	2.16
Packed humid aging	9.5	2.42

TABLE 5. Tests of Treatment Effects on Aerodynamic Size Distribution (Air Velocity in Wind Tunnel = 0.9 cm/sec)

Test Number	Description	S1, Sandy loam soil			S3, clay soil		
		F, statistic	Degrees of freedom n_1, n_2	P probability	F, statistic	Degrees of freedom n_1, n_2	P probability
1.	Overall difference between treatment regimens	0.81	5,146	0.66	4.4	5,128	< 0.0001
2.	Loose dry aging V.S. loose humid aging	1.05	2,94	0.35	0.74	2,77	0.48
3.	Loose aging V.S. packed dry aging	0.37	2,114	0.69	0.57	2,98	0.57
4.	Loose aging V.S. packed humid aging	0.82	2,135	0.44	17.4	2,117	< 0.0001

The overall aerodynamic diameter distribution of $^{238}PuO_2$ on clay soil was larger (AMAD = 9.5 μm, GSD = 2.2) than the aerodynamic diameter distribution of $^{238}PuO_2$ on the sandy loam soil (AMAD = 4.8 μm, GSD = 1.8). This distribution for the sandy loam soil/plutonium aerosol was smaller than the aerodynamic diameter mass distribution of the sandy loam soil alone (MMAD = 5.05 μm, GSD = 2.30). In contrast, the activity distribution for the clay soil/plutonium aerosol was larger than the aerodynamic diameter mass distribution of the clay soil alone (MMAD = 3.35 μm, GSD = 2.45).

The autoradiographic samples were analyzed by graphical comparison of the cumulative distribution curves. In all but one treatment, there were no clear differences among the cumulative distribution curves, even though the parameters of the bimodal distribution indicated that such differences might exist (Table 3). In the sedimentation samples for the clay soil, a significant difference appeared: The size distribution of the attached plutonium particles shifted after humid aging. The count median diameter of the large particles increased, by a factor of four, to 0.8 μm.

The experimental system was fairly reproducible: There was remarkable agreement between the total amount generated as calculated from the sampled data and the data from the generator output (Table 2). However, several cascade impactor samples gave nonsense results (for example, runs 56-12 and 56-41, Table 2). Although "hot" particles[7] were not seen in the autoradiographic samples from this experiment, their presence could explain our results.

In this series of experiments we set up the aerosol generation to model a worst case of resuspension in soil saltation.[4] We had expected to duplicate exposure to rain, but water droplets made the clay soil "bead" into hard balls that could not easily be generated after drying. Humidity cycling was chosen as a best estimate of moisture variations which might be experienced by a

mixture of soil and $^{238}PuO_2$. The soil/plutonium mixture was modeled after an estimated release of 252 Ci of fractured $^{238}PuO_2$ fuel due to impact of a power source with soil.[30] If this amount of fractured $^{238}PuO_2$ fuel were uniformily mixed with the top 5 cm of soil in a circle 1000 m in diameter (about a half-mile), and allowed to stand undisturbed, the soil concentrations would be similar to those seen at the Rocky Flats site[4,7] and the airborne concentration, calculated from the redispersion factor[10] after 5 years (equation 1), would be approximately equal to the MPC (0.6 pCi/m^3). The contaminated area at the Nevada test site covered a circle approximately 300 m in diameter.[5]

Hayden,[3] Tamura,[10,11] and Gillette[15] suggest that if more energy is expended in disrupting the soil to produce an aerosol, a greater fraction of the attached plutonium will be dissociated from the larger particles. Although our method of aerosol generation accelerated all material scraped from the surface of the soil cylinder into a turbulent air stream, increased dissociation of respirable particles was not seen. About 5 to 10% of mixed sandy loam soil and plutonium became airborne with soil, a percentage similar to that seen in saltation from sand in a windstorm.[3] About 15% of the total $^{238}PuO_2$ activity was associated with the 20% of the total mass of clay soil airborne at the end of the wind tunnel. The inverse relationship,[6] between the humidity aging treatment and the total resuspended mass, was not clearly evident.

SUMMARY

Little change in the respirable fraction of airborne particles produced by mechanical disturbance of soil occurred under different aging conditions. Only a slight change was seen in the airborne distribution for clay soil/plutonium mixture under humid conditions (Tables 2 and 3). Humidity aging caused the $^{238}PuO_2$ particles to be more uniformily distributed on the surface of the resuspended clay particles; however, the change was too small to include in safety factor calculations. Other reports[4,5,8,13] show that resuspended mass (activity) above a site will reach safe levels for continuous exposures in as little as 5 years. Therefore reduction in total resuspended mass concentration, rather than the change in activity distribution of resuspended particles, is a more useful variable in predicting change in the inhalation hazard above a site of mixed plutonium and soil.

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